

## Valuing the air quality effects of biochar reductions on soil NO emissions

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## Abstract

While it is clear that biochar can alter soil  $\text{N}_2\text{O}$  emissions, data on NO impacts are scarce. Reports range from 0-67% soil NO emission reductions post-biochar amendment. We use regional air quality and health cost models to assess how these soil NO reductions could influence U.S. air quality and health costs. We find that at 67% soil NO reduction, widespread application of biochar to fertilized agricultural soils could reduce  $\text{O}_3$  by up to 2.4ppb and  $\text{PM}_{2.5}$  by up to  $0.15\mu\text{g}/\text{m}^3$  in some regions. Modeled biochar-mediated health benefits are up to \$4.3 million/county in 2011, with impacts focused in the Midwest and Southwest. These potential air quality and health co-benefits of biochar use highlight the need for an improved understanding of biochar's impacts on soil NO emissions. The benefits reported here should be included with estimates of other biochar benefits, such as crop yield increase, soil water management, and  $\text{N}_2\text{O}$  reductions.

## ■ Introduction

Biochar is intentionally produced charcoal, made through low oxygen heating of organic materials. Biochar soil amendment sequesters carbon and can sometimes improve agricultural productivity<sup>1-3</sup>. Biochar's properties such as high porosity, high surface area and high cation exchange capacity have generated interest in other benefits it may offer<sup>4</sup>. One relevant ancillary benefit is biochar's influence on nitrogen dynamics in fertilized soils<sup>5</sup>, which are major sources of nitrous oxide (N<sub>2</sub>O) and nitric oxide (NO) to the atmosphere<sup>6</sup>. N<sub>2</sub>O is a potent greenhouse gas that contributes to depletion of the stratospheric ozone layer, and NO contributes to the formation of local ozone (O<sub>3</sub>) and fine particulate matter (PM<sub>2.5</sub>)<sup>7-9</sup>. U.S. National Ambient Air Quality Standards (NAAQS) set maximum concentrations of 70 parts per billion (ppb) for the fourth highest 8-hour daily average (MDA8) O<sub>3</sub> and 12µg/m<sup>3</sup> for annual mean PM<sub>2.5</sub><sup>10</sup>. Exposure to O<sub>3</sub> and PM<sub>2.5</sub> is associated with increased risks of premature morbidity and mortality<sup>11-13</sup>, which carry considerable societal costs<sup>14-16</sup>.

Soil biological processes (mainly nitrification and denitrification) are the major controls on soil emissions of NO and N<sub>2</sub>O and the extent of biochar's impact on these processes varies with factors such as soil pH<sup>17, 18</sup> or water content<sup>19</sup>. Although biochar's impact on soil N<sub>2</sub>O has been extensively investigated<sup>20</sup>, its effect on soil NO fluxes has been far less studied<sup>17</sup>. These limited biochar soil NO emission studies have yielded variable results, suggesting that biochar's potential for reducing agricultural air pollution remains poorly constrained. Measurements of biochar's impact on soil NO emissions range from nearly no effect<sup>21</sup> to up to 67% reduction in NO emission from fertilized soils<sup>22</sup>. While this range is large, it is possible to use these values (0-67% reduction in soil NO

emissions) to make a first estimate of the magnitude of potential air quality improvements and reduced health risks associated with biochar application.

The economic returns due to potential biochar-mediated air quality and health benefits have not yet been considered in the cost-benefit analysis of biochar production. Previous studies focused mainly on biochar's production costs and farmers' profit from increased productivity with biochar application<sup>23-25</sup>, but studies have not yet addressed the benefits from mitigating various environmental externalities. Here, we demonstrated an approach to monetizing the air quality benefits associated with agricultural biochar application.

Our approach integrated three models: a soil NO emissions model<sup>26</sup>, an air quality model<sup>8</sup>, and a health cost model<sup>27</sup>. First, we modeled the soil NO reductions resulting from biochar application to U.S. agricultural soils. Next, we used an air quality model to evaluate the subsequent changes in O<sub>3</sub> and PM<sub>2.5</sub>. Then we modeled the health care cost savings of this strategy for local communities across the U.S. over one year. We identified locations where biochar could have the greatest impacts on air quality and health. Our work highlights two points: 1) the potential scale of biochar's air quality and health benefits, and 2) the need for more data on biochar-driven changes in soil NO emissions.

## ■ Methodology

### Goal and Scope

We use the results of the existing biochar soil NO studies as boundary conditions in evaluating biochar's effect on local air quality. While Xiang et al<sup>21</sup> observed an

insignificant change of soil NO emission in a rice-wheat rotation system, a study by Obia et al.<sup>17</sup> of rice husk and cacao shell-derived biochars in fertilized, acidic, sandy loam soil demonstrated a suppression of net NO production and a reduction of its peak over a broad range. Another study by Nelissen et al.<sup>22</sup> showed that amendment of woody and crop-based biochars to silt loam soil in a primarily nitrogenous fertilized environment has resulted in 47% to 67% reduction in NO emission. The variation in results reported by these studies is consistent with the notion that biochar's effects on soil microbial processes may be specific to biochar chemical properties (e.g. biochar pH) and/or physical properties<sup>28, 29</sup>, driven by its biomass of origin, by production process, biochar's C/N ratio, and by the effects of environmental aging. These factors, in addition to changes in meteorological conditions, have been shown to alter nitrogenous gas emissions from soils amended with biochar, including N<sub>2</sub>O<sup>30, 31</sup> primarily by limiting soil nitrogen availability and altering the N<sub>2</sub>O product ratios of both nitrification and denitrification<sup>18</sup>. Because NO is also a product of nitrification and denitrification<sup>32</sup>, we expect that NO emission from soil amended with biochar will also undergo changes in response to meteorological conditions. We use the range of 0% to 67% NO reduction for lower and upper limits in our study, given that the few available studies present a broad range of changes in soil NO emission affected by biochar presence. We also test a scenario where biochar amendment induces a 47% reduction in soil emission to demonstrate how a different NO reduction value may affect the health benefit estimates of the study (supporting information).

To estimate agricultural soil NO emission with and without biochar, we apply an advanced parametrization method that includes key processes and parameters to

represent available nitrogen in the soil. We run this model under two conditions: 1) base condition (no biochar application, or 0% NO reduction), and 2) biochar application (67% NO reduction, consistent with the upper value reported in the current literature<sup>22</sup>).

We model altered soil NO emissions for currently fertilized agricultural soils in the continental US. Soil NO emissions are estimated for two time periods – one month (July 2011) and one full year (2011) as a representative baseline. July NO emission changes are used in a photochemical air quality model to simulate changes in air quality. In a given year, July tends to be a month of peak O<sub>3</sub> concentrations and is a month when NO emissions from fertilized fields can be relatively large. The NO emission changes results we base our analysis on were also measured immediately after fertilization event (14 and 8 consecutive days), where majority of fertilizer-related NO emission takes place<sup>17, 22</sup>. Running the air quality model based on July NO results allows testing the effectiveness of biochar in O<sub>3</sub> and PM<sub>2.5</sub> standard attainments of local governments. To estimate the annual soil NO changes, our model accounts for timing, frequency and spatial variation of the fertilizer application across agricultural regions in the U.S. The annual soil NO emission changes are then used in a health effects valuation model to associate those NO emission changes with their long-term impacts on O<sub>3</sub> and PM<sub>2.5</sub> emission changes and human health. While a previous study has documented an increased risk of PM<sub>10</sub> emissions from soil biochar amendment<sup>33</sup>, we focus on PM<sub>2.5</sub> reduction because: 1) the small sizes of PM<sub>2.5</sub> particles cause a stronger risk to health (especially mortality) than coarser parts of PM<sub>10</sub>, and 2) we assume that biochar users will follow the best practices in biochar application and the International Biochar

Initiative (IBI) guidelines in maintaining a minimum biochar moisture level to minimize the biochar wind erosion<sup>34, 35</sup>.

The following sections present detailed descriptions of the models used.

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### 111 **Soil NO estimates and O<sub>3</sub> and PM<sub>2.5</sub> concentration changes**

The Berkeley-Dalhousie Soil NO<sub>x</sub> Parameterization (BDSNP) estimates NO emissions as a function of nitrogen availability, soil temperature, soil moisture, and other factors<sup>26</sup>.

We apply BDSNP in two ways – an inline version incorporated into the Community Multiscale Air Quality (CMAQ) model<sup>8</sup>, and a less computationally intensive offline version.

Each version of BDSNP estimates soil NO based on a biome-specific base emission factor ( $A'_{biome}$ ) and an available soil nitrogen pool ( $N_{avail}$ ) originating from fertilizer application and nitrogen deposition from the atmosphere. Emission rates are modulated based on response functions to soil temperature ( $f(T)$ ) and soil moisture( $g(\theta)$ ), a soil pulsing factor ( $P$ ) when precipitation follows a dry period ( $l_{dry}$ ), and a canopy reduction factor (CRF) that depends on biome type, leaf area index, and meteorology.

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$$124 \quad \text{Soil NO Flux} = A'_{biome}(N_{avail}) \times f(T) \times g(\theta) \times P(l_{dry}) \times CRF \quad (1)$$

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The inline version computes  $N_{avail}$  based on nitrogen deposition computed within CMAQ, while the offline version takes deposition fields from an archived CMAQ run.

Each version uses the global fertilizer database from Potter et al.<sup>36</sup> and assumes that 37% of fertilizer and manure N is available for potential emission<sup>26</sup>. Biome-specific base emission factors are taken from Steinkamp and Lawrence<sup>37</sup> using Köppen climate zone classifications<sup>38</sup>, as described by Rasool et al.<sup>8</sup>.

We apply the inline version of CMAQ-BDSNP to simulate one month of the growing season (July 2011), and offline BDSNP to simulate full year 2011. CMAQ runs compute the changes in O<sub>3</sub> and PM<sub>2.5</sub> concentrations in July 2011 under two different scenarios – one without biochar and another with biochar soil amendment. The models are applied over a domain covering the continental US (CONUS) with horizontal resolution of 12 km x 12 km.

Meteorological fields influencing atmospheric and soil conditions are taken from a simulation with the Weather Research and Forecasting (WRF) model<sup>39</sup>. Model configurations for WRF, BDSNP, and CMAQ are summarized in Table 1.

In addition, we ran the stand-alone BDSNP in July for five years (2009, 2010, 2011, 2014 and 2015) with different El Niño/Southern Oscillation (ENSO) index conditions (wet or dry year) to bound the variation range of possible soil NO emission reduction due to biochar. The results suggest the bias for July 2011 modeling outputs is within ± 20% under different meteorological conditions (for more details refer to supporting information).



149 **Table 1. Configuration of the WRF-BDSNP-CMAQ model used in this study**

WRF/MCIP			
Version	ARW V3.7	Shortwave radiation	RRTMG scheme
Horizontal resolution	CONUS (12 km x 12 km)	Surface layer physics	Pleim-Xiu surface model
Vertical resolution	26 layers	PBL Scheme	ACM2
Boundary condition	NARR 32 km	Microphysics	Morrison double-moment scheme
Initial condition	NCEP-ADP	Cumulus parameterization	Kain-Fritsch scheme
Longwave radiation	RRTMG scheme	Assimilator	Analysis nudging above PBL for temperature, moisture and wind speed
BDSNP			
Horizontal resolution	Same as WRF/MCIP	Emission factor	Steinkamp and Lawrence (2011)
Soil Biome type	24 types based on NLCD40	Fertilizer database	Potter et al. (2010)
CMAQ			
Version	V5.0.2	Anthropogenic emission	NEI2011
Horizontal resolution	Same as WRF/MCIP	Biogenic emission	BEIS V3.14 inline
Initial condition	GEOS-Chem	Boundary condition	GEOS-Chem
Aerosol module	AE5	Gas-phase mechanism	CB-05
Simulation Case Arrangement			
Control	WRF-BDSNP-CMAQ simulation with standard configuration		
Biochar	WRF-BDSNP-CMAQ simulation with the soil NO emission scaled down by 67% over the regions with N fertilizer application		
Simulation Time Period			
WRF-BDSNP (inline)-CMAQ	July 1-30, 2011 for CMAQ simulation with inline soil NO BDSNP module		
WRF-BDSNP (standalone)-AP2	Full year (2011)		
WRF-BDSNP (standalone)	Time of maximum fertilizer application i.e. July in regions of dominant fertilizer application compared for 5 different years based on ENSO index: 2009 (0.6, modest El Nino year); 2010 (-1.1, Strong La Niña year); 2011 (-0.5, modest La Niña year); 2014 (0, normal year) and 2015 (1.5; strong El Niño year)		

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## **Estimating the health impact costs of changes in air pollutants through concentration-response functions**

We use the AP2 model<sup>27</sup>, an updated version of the Air Pollution Emission Experiments and Policy Analysis (APEEP) model<sup>40</sup>, to evaluate the health impacts of reduced air pollution. The cost module from AP2 uses concentration-response (C-R) functions from epidemiological studies, which indicate the susceptibility of population age groups to relate changes in  $O_3$ <sup>11, 41-43</sup> and  $PM_{2.5}$ <sup>13, 44, 45</sup> concentrations to morbidity and mortality. Impacts on morbidity and mortality rates of local communities are quantified by associating the air pollution changes and C-R functions with county-level demographic profiles.

In comparison to US EPA values, AP2 estimates lower health care savings associated with air quality changes<sup>27</sup>. Aside from differences in their air quality models, a lower value of statistical life (VSL) assumption in cost module of AP2 may contribute to lower damage estimates by this model<sup>46</sup>. In our study we update the morbidity willingness to pay (WTP) values in AP2 with discounted 2011 values (Table 2) and replace mortality cost with the EPA's VSL reported for 2011<sup>47</sup>. US Census data<sup>48</sup> for 2011 is used to update the AP2 county-level population input.

We run AP2 for a baseline condition and a second scenario where biochar reduces annual emissions of NO in fertilized agricultural soil in 2011. The differences in estimated damages from these runs demonstrate the potential annual health care savings of biochar-mediated NO emissions.

**Table 2. Unit values used for VSL and WTP, 2011 dollars**

Basis for estimate	Age range	Unit value
VSL	0 - 99	\$8,200,000 <sup>1</sup>
WTP for Asthma	30 - 99	\$42,593 <sup>2</sup>
WTP for Chronic Bronchitis	30 - 99	\$442,955 <sup>3</sup>

<sup>1</sup> US EPA<sup>47</sup>

<sup>2</sup> AP2 WTP for asthma updated for 2011\$ with a 3% discount rate

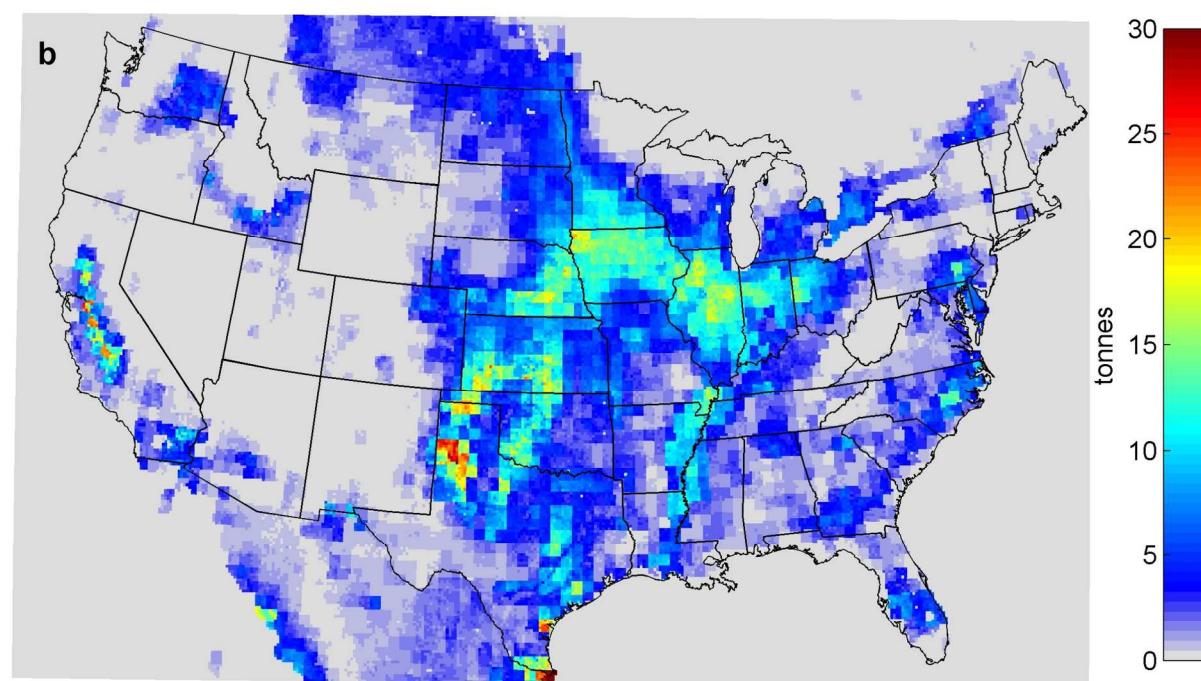
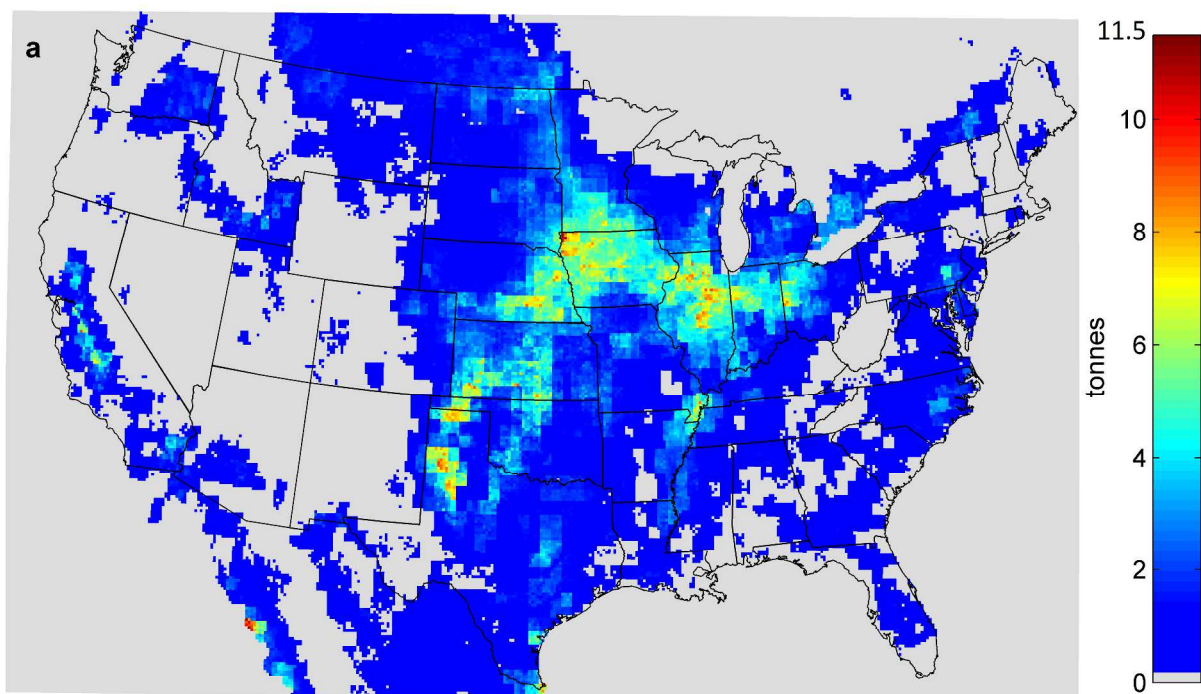
<sup>3</sup> AP2 WTP for chronic bronchitis updated for 2011\$ with a 3% discount rate

**■ Results and Discussion**

This section describes the effect of biochar application if it reduces 67% of fertilized soil NO emission. In addition, we have tested a scenario of 47% NO reduction for a point of comparison, which is presented in supporting information (Figures 1S to 3S).

**Changes in seasonal and annual NO emissions across US**

We used the offline BDSNP model to estimate a total base soil NO emission of 648,000 tonnes/year in 2011 (Table S1). Then we estimated the reduction in NO emissions that would have occurred in 2011 if biochar had reduced emissions from fertilized agricultural soils by 67%, while emissions from other soils remain unchanged (Figure 1). The greatest reductions in soil NO emission on a percentage basis occur in states with large amounts of fertilized soils such as Kansas (-33.5%), Idaho (-30.0%), Ohio (-8.4%), and Iowa (-18.4%). Application of biochar to fertilized soils across the continental US would reduce soil NO emissions by 90,000 tonnes/year or -12.3% (Table S1), based on the upper level values reported by Nelissen et al.<sup>22</sup>.



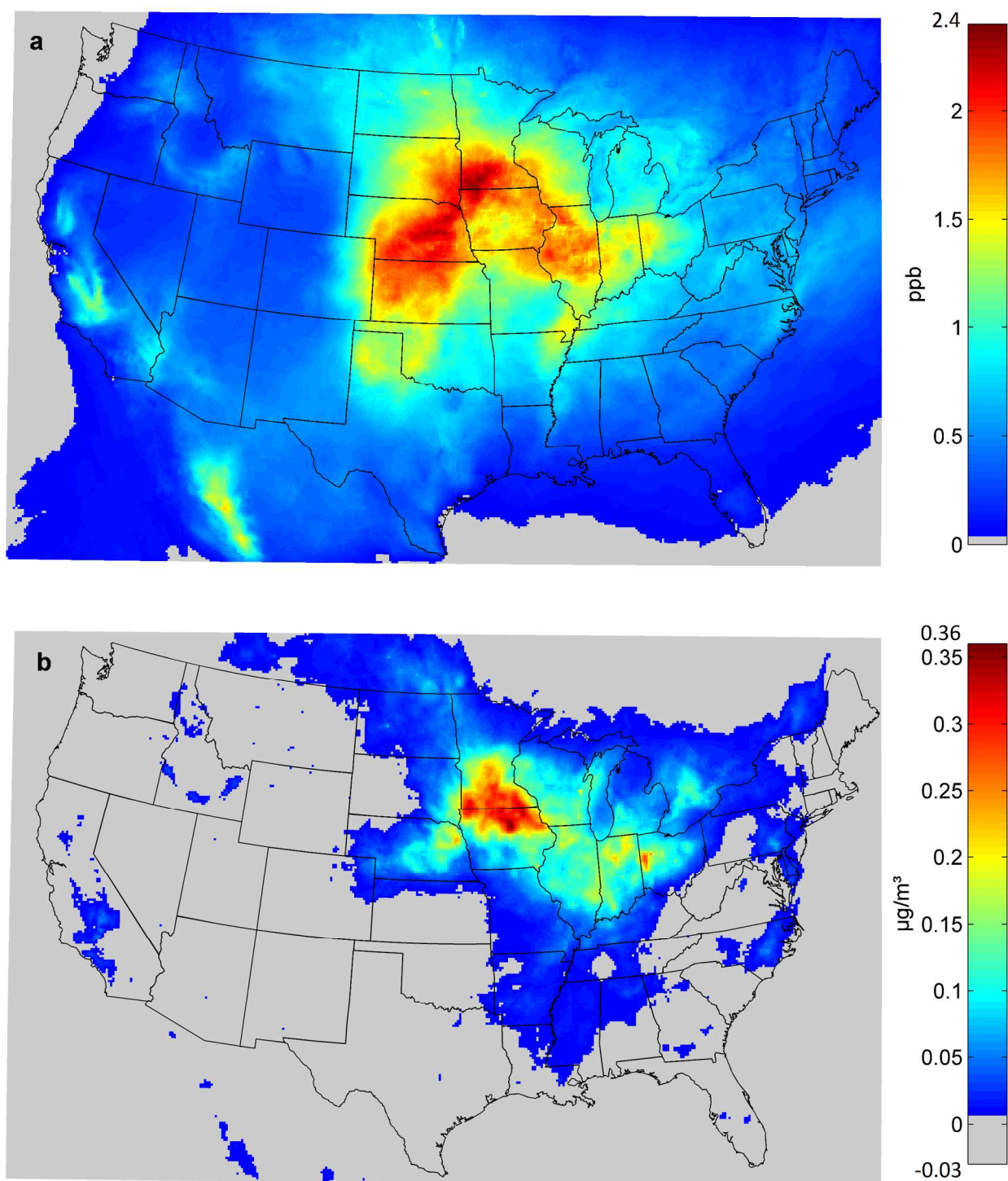
**Figure 1. Reductions in soil NO in a) July 2011, and b) 2011, if biochar application reduces fertilized soil NO emissions by 67%. Map credit: MATLAB and Mapping**

**Toolbox Release 2014a., The MathWorks, Inc., Natick, Massachusetts, United States.**

### **Changes in seasonal O<sub>3</sub> and PM<sub>2.5</sub> concentrations across US**

Running the CMAQ simulations showed reductions in MDA8 O<sub>3</sub> of at least 1ppb for much of the Midwest and San Joaquin Valley, with a maximum impact of 2.4ppb (Figures 2a). These regions had the largest soil NO emissions from fertilized agriculture (Figure 1) and tend to have NO<sub>x</sub>-limited O<sub>3</sub> formation<sup>49, 50</sup> (NO<sub>x</sub>=NO and NO<sub>2</sub>). The reduction of 1-2ppb can have meaningful implications mainly for urban areas near agricultural areas that require small O<sub>3</sub> reductions to comply with the 70ppb standard<sup>51</sup>. For example, as of 2013-2015 data, Chicago, St. Louis, Cleveland, Columbus, and Cincinnati all have MDA8 O<sub>3</sub> levels of 1-5ppb above the standard<sup>52</sup>. Thus, reducing NO emission from agricultural soils in these locations can be considered a plausible component of their portfolio to manage O<sub>3</sub> levels.

For PM<sub>2.5</sub>, biochar application reduced concentrations by more than 0.1µg/m<sup>3</sup> over portions of the Midwest in July 2011, with a maximum impact of 0.33µg/m<sup>3</sup> (Figure 2b). The decline in PM<sub>2.5</sub> levels occur via reductions in ammonium nitrate aerosol formation, and because NO plays a minor role in influencing rates of formation of secondary organic aerosols. However, PM<sub>2.5</sub> is modeled to slightly increase (< 0.03µg/m<sup>3</sup>) in sulfate-rich regions where nitrate competes with sulfate to react with ammonium to form PM<sub>2.5</sub><sup>53</sup>.



**Figure 2. Absolute change in July monthly mean of a) MDA8 O<sub>3</sub> (ppb), and b) PM<sub>2.5</sub> (µg/m<sup>3</sup>), if biochar application reduces fertilized soil NO emissions by 67%.**

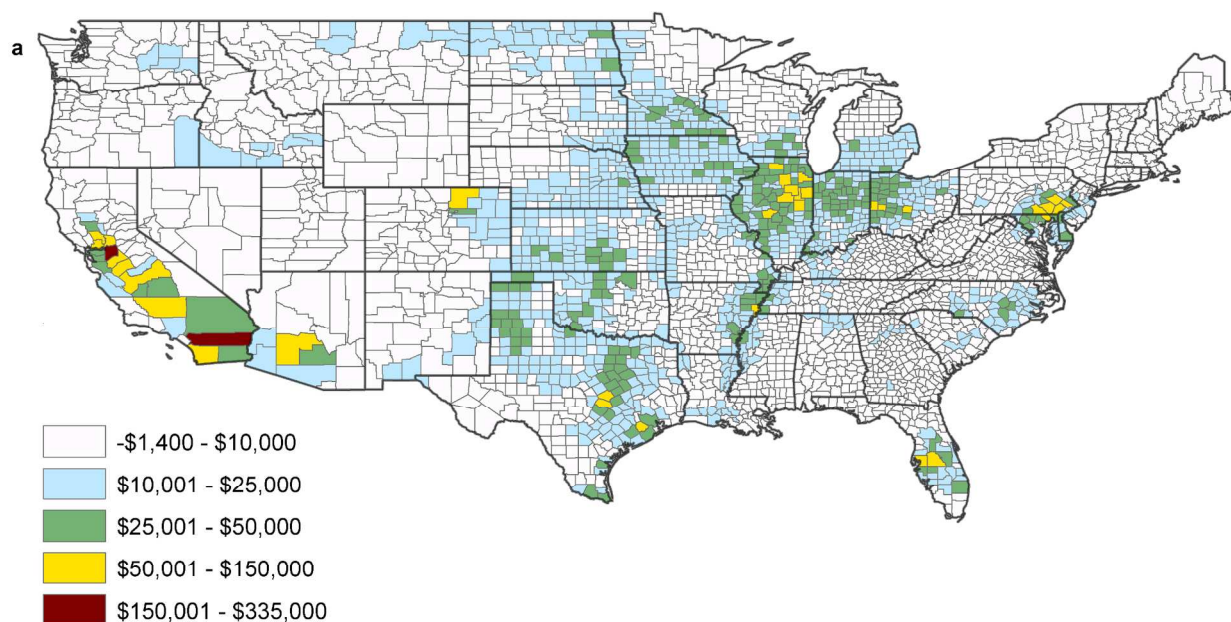
**Potential health benefits of biochar: regions close to populous cities receive the highest health benefits of biochar application. Map credit: MATLAB and Mapping Toolbox Release 2014a., The MathWorks, Inc., Natick, Massachusetts, United States.**

The economic model used here ( AP2<sup>27</sup>) predicted that nationwide application of biochar to agricultural soils with 67% reduction of their annual NO emissions would reduce \$660 million of the health impacts of agricultural air pollution for the entire US. Changes in O<sub>3</sub> concentration reduced agricultural health impacts by up to \$335,000/county and improvements in PM<sub>2.5</sub> concentrations would result in health benefits of up to \$4.2 million/county. The median county-level health savings of O<sub>3</sub> and PM<sub>2.5</sub> reductions were \$6000 and \$97,000 across regions of the US with agricultural activities. These results contain considerable spatial variation, with some regions seeing significant benefits, and others none. While many agricultural areas across the US showed higher health cost savings, the largest benefits occur in areas such as California's Central Valley (Figure 3) (see the ranking details of county-level savings in Tables S2 and S3). For example, a few counties in California's Central Valley saw savings of more than \$2 million (e.g. Fresno County). Of course, total dollar values should be viewed with caution, because they depend on assumptions about the Value of a Statistical Life (VSL) and the impact of biochar application on soil NO emissions. Nevertheless, the results of the analysis indicate which regions are likely to see the greatest relative impact, which makes them informative to policymakers, the agricultural community and the public on the potential local air quality benefits of biochar application.

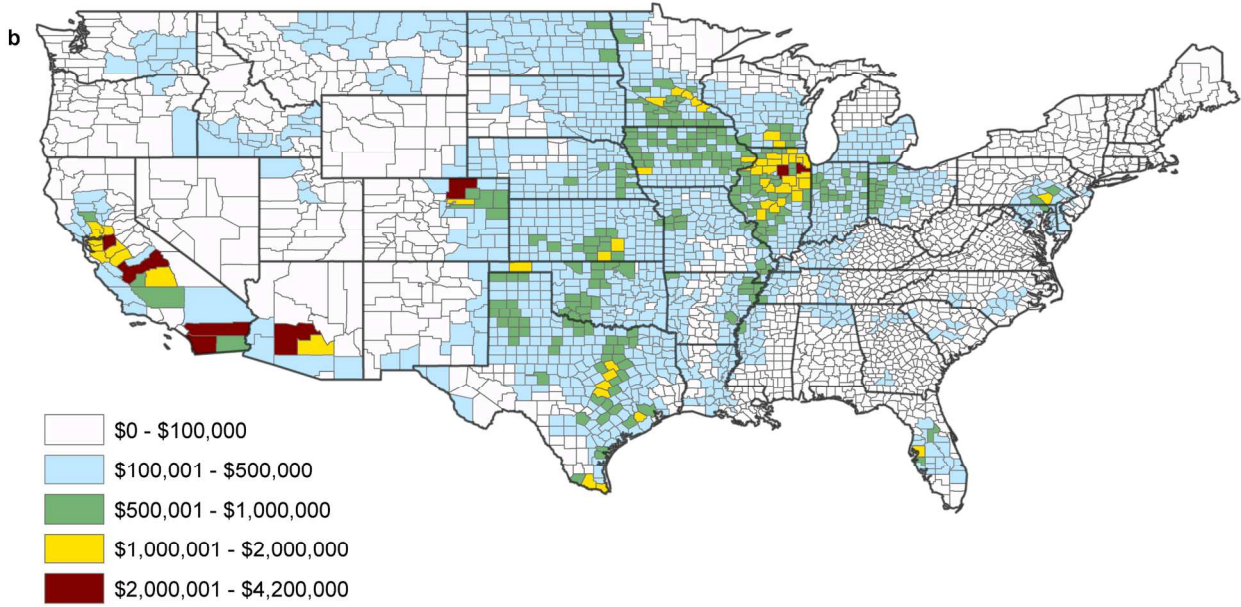


There are two primary factors influencing the estimated regional health benefits – the level of agricultural activity and demography. In particular, small reductions in  $O_3$  or  $PM_{2.5}$  resulting from biochar application in agricultural settings near highly populous areas result in larger savings (compare Figures 1 and 3). Indeed, our results show that the intersection of high agricultural  $NO$  reduction upwind of densely populated regions drives health benefits from agricultural  $NO$  emissions reductions. This can be seen most clearly in California and Illinois.

The savings through reducing  $PM_{2.5}$  were almost 10x larger than those from  $O_3$  reductions (Figure 3a vs 3b). This is because  $PM_{2.5}$  has more potent health impacts than  $O_3$ . These simulation results show substantial opportunities for reducing health costs that are caused by agricultural activities near populous cities, in particular, in the mid and upper Midwest and California<sup>54</sup>.







**Figure 3. County-level health cost savings due to a) O<sub>3</sub> reduction, and b) PM<sub>2.5</sub> reduction, if biochar application reduces fertilized soil NO emissions by 67%. Map credit: USA Counties, ArcGIS 10.3, ESRI, Tom Tom, U.S. Department of Commerce, U.S. Census Bureau.**

**Improving local air quality: an added biochar benefit**

Agriculture is a major source of ecosystem pollution and is responsible for up to one-fifth of air pollution mortality globally<sup>55</sup>. Through several programs and incentives<sup>56, 57</sup>, the U.S. has been promoting farming practices that mitigate agricultural air quality issues. Reducing NO emission from agricultural soils<sup>58</sup> may benefit air quality and health. However, soil NO emissions have largely been ignored in state strategies for attaining O<sub>3</sub> and PM<sub>2.5</sub> standards, representing an untapped opportunity for mitigation. If the Nelissen<sup>22</sup> findings of biochar impact on soil NO emissions are representative nationally, soil application of biochar may yield tangible air quality and health benefits in

regions of the U.S. struggling with agriculture-related smog. Our results show that biochar soil application may reduce the emission of up to 90,000 tonnes of NO from the US agricultural sector during the year of application. Although NO reduction benefits are experienced locally, we estimated that nationwide application of biochar could yield \$660 million in health benefits if biochar indeed reduces fertilized soil NO by 67%. Thus, these results make clear the urgent need for analyses of biochar-influenced soil NO changes to better understand the air quality value of biochar in agricultural soils.

Our study helps to identify areas where more information is needed to validate biochar performance in reducing soil NO emission. Spatial patterns of reduction in emissions of NO (Figure 1) and concentrations of O<sub>3</sub> and PM<sub>2.5</sub> (Figure. 2) can be used as a guide to prioritizing locations for further study or deployment of biochar. California, Arizona, and the Midwest are most likely to benefit from reductions in agricultural NO emissions (Figure 3).

Our analysis demonstrates that there may be a positive value associated with biochar application, but realizing that value may require policy incentives that allow monetization of NO reduction. Importantly, well-designed policy could stimulate market valuation of the avoided externalities associated with biochar application in agricultural soils.

Biochar's potential in achieving health benefits through improved air quality is an additional value complementing biochar's other services (e.g. crop yield improvement and reduction in nutrient pollution through retention of N and P within agroecosystems). Ultimately, decision-making for implementing biochar in a location will require a full analysis of all biochar's variable benefits as well as the costs involved that are also dependent on region, feedstock and process design and crop choice. Hence,

quantifying the avoided externalities associated with biochar application, and designing market mechanisms to dictate the value of biochar's potential ecosystem services, help to better evaluate the opportunities associated with large-scale application of biochar. As such, policy can play an important role in achieving these benefits and lead to an increased use of biochar.

Before efficient policy design can be undertaken, however, certain areas require deeper investigation. In particular, we need: 1) a better understanding of biochar's short and long-term influence on soil nitrogen dynamics under a variety of meteorological conditions, specifically emission of air pollutant precursors like NO, and 2) improvement in biogeochemical models or soil NO parametrization schemes that simulate biochar presence in soil and linking their simulated results to regional air quality models. Integration of such improved measurements with cost models that estimate the regional impacts of agricultural practices will help local communities make informed policy decisions regarding agricultural practices and local air quality management.

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## 311 **Acknowledgement**

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**Supporting Information.** Maps of O<sub>3</sub> and PM<sub>2.5</sub>, and health benefits due to 47% fertilized soil NO reduction when biochar is applied, Tables of State level NO reductions and County level health benefits

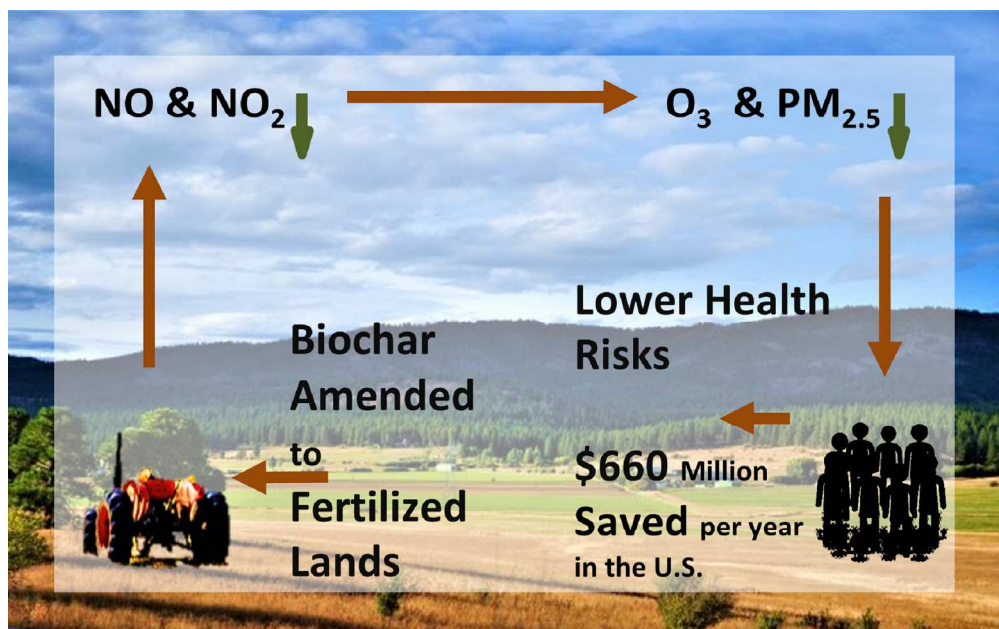
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319 **References**

- 320 1. Lehmann, J., A handful of carbon. *Nature* **2007**, *447*, (7141), 143-144.
- 321 2. Marris, E., Putting the carbon back: Black is the new green. *Nature* **2006**, *442*, (7103), 624-626.
- 322 3. Lehmann, J.; Pereira da Silva, J.; Steiner, C.; Nehls, T.; Zech, W.; Glaser, B., Nutrient availability  
323 and leaching in an archaeological Anthrosol and a Ferralsol of the Central Amazon basin: fertilizer,  
324 manure and charcoal amendments. *Plant Soil* **2003**, *249*, (2), 343-357.
- 325 4. Sohi, S. P.; Krull, E.; Lopez-Capel, E.; Bol, R., Chapter 2 - A Review of Biochar and Its Use and  
326 Function in Soil. In *Adv. Agron.*, Academic Press: 2010; Vol. Volume 105, pp 47-82.
- 327 5. Clough, T. J.; Condon, L. M., Biochar and the Nitrogen Cycle: Introduction *J. Environ. Qual.* **2010**,  
328 (4), 1218-1223.
- 329 6. Bouwman, A. F.; Boumans, L. J. M.; Batjes, N. H., Emissions of N<sub>2</sub>O and NO from fertilized fields:  
330 Summary of available measurement data. *Global Biogeochem. Cycles* **2002**, *16*, (4), 6-1-6-13.
- 331 7. Seinfeld, J. H.; Pandis, S. N., *Atmospheric Chemistry and Physics: From Air Pollution to Climate*  
332 *Change*. 2nd ed.; 1998.
- 333 8. Rasool, Q. Z.; Zhang, R.; Lash, B.; Cohan, D. S.; Cooter, E. J.; Bash, J. O.; Lamsal, L. N., Enhanced  
334 representation of soil NO emissions in the Community Multiscale Air Quality (CMAQ) model  
335 version 5.0.2. *Geosci. Model Dev.* **2016**, *9*, (9), 3177-3197.
- 336 9. Peel, J. L.; Haeuber, R.; Garcia, V.; Russell, A. G.; Neas, L., Impact of nitrogen and climate change  
337 interactions on ambient air pollution and human health. *Biogeochemistry* **2013**, *114*, (1), 121-134.
- 338 10. EPA NAAQS Table. <https://www.epa.gov/criteria-air-pollutants/naaqs-table> (June 7),
- 339 11. Bell, M. L.; McDermott, A.; Zeger, S. L.; Samet, J. M.; Dominici, F., Ozone and short-term  
340 mortality in 95 us urban communities, 1987-2000. *JAMA* **2004**, *292*, (19), 2372-2378.
- 341 12. Jerrett, M.; Burnett, R. T.; Pope, C. A.; Ito, K.; Thurston, G.; Krewski, D.; Shi, Y.; Calle, E.; Thun,  
342 M., Long-Term Ozone Exposure and Mortality. *N. Engl. J. Med.* **2009**, *360*, (11), 1085-1095.
- 343 13. Pope, C. A.; Burnett, R. T.; Thun, M. J.; Calle, E. E.; Krewski, D.; Ito, K.; Thurston, G. D., Lung  
344 Cancer, Cardiopulmonary mortality, and long-exposure to fine particulate air pollution. *JAMA* **2002**, *287*.
- 345 14. Li, J.; Ewart, G.; Kraft, M.; Finn, P. W., The public health benefits of air pollution control. *J.*  
346 *Allergy Clin. Immunol.* **2012**, *130*, (1), 22-23.
- 347 15. Pervin, T.; Gerdtham, U.-G.; Lyttkens, C. H., Societal costs of air pollution-related health hazards:  
348 A review of methods and results. *Cost Effectiveness and Resource Allocation* **2008**, *6*, (1), 1-22.
- 349 16. Landrigan, P. J., The hidden costs of environmental contamination. *Eur. Respir. J.* **2012**, *40*, (2),  
350 286-288.
- 351 17. Obia, A.; Cornelissen, G.; Mulder, J.; Dörsch, P., Effect of Soil pH Increase by Biochar on NO,  
352 N(2)O and N(2) Production during Denitrification in Acid Soils. *PLoS ONE* **2015**, *10*, (9), e0138781.
- 353 18. Mørkvad, P. T.; Dörsch, P.; Bakken, L. R., The N<sub>2</sub>O product ratio of nitrification and its  
354 dependence on long-term changes in soil pH. *Soil Biol. Biochem.* **2007**, *39*, (8), 2048-2057.
- 355 19. Saarnio, S.; Heimonen, K.; Kettunen, R., Biochar addition indirectly affects N<sub>2</sub>O emissions via soil  
356 moisture and plant N uptake. *Soil Biol. Biochem.* **2013**, *58*, 99-106.
- 357 20. Cayuela, M. L.; van Zwieten, L.; Singh, B. P.; Jeffery, S.; Roig, A.; Sánchez-Monedero, M. A.,  
358 Biochar's role in mitigating soil nitrous oxide emissions: A review and meta-analysis. *Agric., Ecosyst.*  
359 *Environ.* **2014**, *191*, 5-16.
- 360 21. Xiang, J.; Liu, D.; Ding, W.; Yuan, J.; Lin, Y., Effects of biochar on nitrous oxide and nitric oxide  
361 emissions from paddy field during the wheat growth season. *J. Cleaner Prod.* **2015**, *104*, 52-58.
- 362 22. Nelissen, V.; Saha, B. K.; Ruysschaert, G.; Boeckx, P., Effect of different biochar and fertilizer  
363 types on N<sub>2</sub>O and NO emissions. *Soil Biol. Biochem.* **2014**, *70*, 244-255.

23. Galinato, S. P.; Yoder, J. K.; Granatstein, D., The economic value of biochar in crop production and carbon sequestration. *Energy Policy* **2011**, 39, (10), 6344-6350.
24. Dickinson, D.; Balduccio, L.; Buysse, J.; Ronsse, F.; van Huylenbroeck, G.; Prins, W., Cost-benefit analysis of using biochar to improve cereals agriculture. *GCB Bioenergy* **2015**, 7, (4), 850-864.
25. Field, J. L.; Keske, C. M. H.; Birch, G. L.; DeFoort, M. W.; Cotrufo, M. F., Distributed biochar and bioenergy coproduction: a regionally specific case study of environmental benefits and economic impacts. *GCB Bioenergy* **2013**, 5, (2), 177-191.
26. Hudman, R. C.; Moore, N. E.; Mebust, A. K.; Martin, R. V.; Russell, A. R.; Valin, L. C.; Cohen, R. C., Steps towards a mechanistic model of global soil nitric oxide emissions: implementation and space based-constraints. *Atmos. Chem. Phys.* **2012**, 12, (16), 7779-7795.
27. Muller, N. Z., Linking policy to statistical uncertainty in air pollution damages. *The B.E. journal of economic analysis & policy* **2011**, 11, (1), 27.
28. Gao, X.; Cheng, H.-Y.; Del Valle, I.; Liu, S.; Masiello, C. A.; Silberg, J. J., Charcoal Disrupts Soil Microbial Communication through a Combination of Signal Sorption and Hydrolysis. *ACS Omega* **2016**, 1, (2), 226-233.
29. Masiello, C. A.; Chen, Y.; Gao, X.; Liu, S.; Cheng, H.-Y.; Bennett, M. R.; Rudgers, J. A.; Wagner, D. S.; Zygourakis, K.; Silberg, J. J., Biochar and Microbial Signaling: Production Conditions Determine Effects on Microbial Communication. *Environ. Sci. Technol.* **2013**, 47, (20), 11496-11503.
30. Spokas, K. A., Impact of biochar field aging on laboratory greenhouse gas production potentials. *GCB Bioenergy* **2013**, 5, (2), 165-176.
31. Hagemann, N.; Harter, J.; Kaldamukova, R.; Guzman-Bustamante, I.; Ruser, R.; Graeff, S.; Kappler, A.; Behrens, S., Does soil aging affect the N<sub>2</sub>O mitigation potential of biochar? A combined microcosm and field study. *GCB Bioenergy* **2017**, 9, (5), 953-964.
32. Parton, W. J.; Holland, E. A.; Del Grosso, S. J.; Hartman, M. D.; Martin, R. E.; Mosier, A. R.; Ojima, D. S.; Schimel, D. S., Generalized model for NO<sub>x</sub> and N<sub>2</sub>O emissions from soils. *J. Geophys. Res.: Atmos.* **2001**, 106, (D15), 17403-17419.
33. Ravi, S.; Sharratt, B. S.; Li, J.; Olshevski, S.; Meng, Z.; Zhang, J., Particulate matter emissions from biochar-amended soils as a potential tradeoff to the negative emission potential. *Scientific Reports* **2016**, 6, 35984.
34. Silva, F. C.; Borrego, C.; Keizer, J. J.; Amorim, J. H.; Verheijen, F. G. A., Effects of moisture content on wind erosion thresholds of biochar. *Atmos. Environ.* **2015**, 123, Part A, 121-128.
35. Major, J. *Guidelines on Practical Aspects of Biochar Application to Field Soil in Various Soil Management Systems*; International Biochar Initiative: 2010.
36. Potter, P.; Ramankutty, N.; Bennett, E. M.; Donner, S. D., Characterizing the Spatial Patterns of Global Fertilizer Application and Manure Production. *Earth Interact.* **2010**, 14, (2), 1-22.
37. Steinkamp, J.; Lawrence, M. G., Improvement and evaluation of simulated global biogenic soil NO emissions in an AC-GCM. *Atmos. Chem. Phys.* **2011**, 11, (12), 6063-6082.
38. Kottek, M.; Grieser, J.; Beck, C.; Rudolf, B.; Rubel, F., World Map of the Köppen-Geiger climate classification updated. *Meteorol. Z.* **2006**, 15, (3), 259-263.
39. Michalakos, J.; Dudhia, J.; Gill, D.; Henderson, T.; Klemp, J.; Skamarock, W.; Wang, W. In *The Weather Research and Forecast Model: Software Architecture and Performance*, 11th ECMWF workshop on the use of high performance computing in meteorology 2005; World Scientific: 2005; pp 156-168.
40. Muller, N. Z.; Mendelsohn, R., Measuring the damages of air pollution in the United States. *Journal of Environmental Economics and Management* **2007**, 54, (1), 1-14.
41. McDonnell, W. F.; Abbey, D. E.; Nishino, N.; Lebowitz, M. D., Long-Term Ambient Ozone Concentration and the Incidence of Asthma in Nonsmoking Adults: The Ahsmog Study. *Environ. Res.* **1999**, 80, (2), 110-121.

42. Stieb, D. M.; Burnett, R. T.; Beveridge, R. C.; Brook, J. R., Association between ozone and asthma emergency department visits in Saint John, New Brunswick, Canada. *Environ. Health Perspect.* **1996**, *104*, (12), 1354-1360.
43. Schwartz, J., Short term fluctuations in air pollution and hospital admissions of the elderly for respiratory disease. *Thorax* **1995**, *50*, (5), 531-538.
44. Woodruff, T. J.; Parker, J. D.; Schoendorf, K. C., Fine Particulate Matter (PM<sub>2.5</sub>) Air Pollution and Selected Causes of Postneonatal Infant Mortality in California. *Environ. Health Perspect.* **2006**, *114*, (5), 786-790.
45. Abbey, D. E.; Ostro, B. E.; Petersen, F.; Burchette, R. J., Chronic respiratory symptoms associated with estimated long-term ambient concentrations of fine particulates less than 2.5 microns in aerodynamic diameter (PM<sub>2.5</sub>) and other air pollutants. *J Expo Anal Environ Epidemiol* **1995**, *5*, (2), 137-59.
46. Fann, N.; Baker, K. R.; Fulcher, C. M., Characterizing the PM<sub>2.5</sub>-related health benefits of emission reductions for 17 industrial, area and mobile emission sectors across the U.S. *Environ. Int.* **2012**, *49*, 141-151.
47. USEPA, Estimating the Benefit per Ton of Reducing PM<sub>2.5</sub> Precursors from 17 Sectors. In Office of Air Quality Planning and Standards: Office of Air and Radiation: 2013.
48. USCensus USA Annual County Population Estimates: April 1, 2010 to July 1, 2015. (July 21),
49. Blanchard, C. L.; Tanenbaum, S., VOC and NO<sub>x</sub> Limitation of Ozone Formation at Monitoring Sites in Illinois, Indiana, Michigan, Missouri, Ohio, and Wisconsin, 1998-2002. **2003**.
50. Pusede, S. E.; Cohen, R. C., On the observed response of ozone to NO<sub>x</sub> and VOC reactivity reductions in San Joaquin Valley California 1995–present. *Atmos. Chem. Phys.* **2012**, *12*, (18), 8323-8339.
51. USEPA, Ozone Designations Mapping Tool V1. In *Web Mapping Application*, 2016.
52. USEPA, 8-Hour Ozone (2008) Designated Area Design Values. In September 22, 2016 ed.; USEPA: 2016.
53. Pinder, R. W.; Dennis, R. L.; Bhave, P. V., Observable indicators of the sensitivity of PM<sub>2.5</sub> nitrate to emission reductions—Part I: Derivation of the adjusted gas ratio and applicability at regulatory-relevant time scales. *Atmos. Environ.* **2008**, *42*, (6), 1275-1286.
54. Oikawa, P. Y.; Ge, C.; Wang, J.; Eberwein, J. R.; Liang, L. L.; Allsman, L. A.; Grantz, D. A.; Jenerette, G. D., Unusually high soil nitrogen oxide emissions influence air quality in a high-temperature agricultural region. *Nat Commun* **2015**, *6*.
55. Lelieveld, J.; Evans, J. S.; Fnais, M.; Giannadaki, D.; Pozzer, A., The contribution of outdoor air pollution sources to premature mortality on a global scale. *Nature* **2015**, *525*, (7569), 367-371.
56. USDA USDA Agricultural Air Quality Task Force.  
<http://www.nrcs.usda.gov/wps/portal/nrcs/main/national/air/taskforce/> (Oct 18),
57. USDA Environmental Quality Incentives Program.  
<http://www.nrcs.usda.gov/wps/portal/nrcs/main/ca/programs/financial/eqip/> (Oct 18),
58. Skiba, U.; Fowler, D.; Smith, K. A., Nitric oxide emissions from agricultural soils in temperate and tropical climates: sources, controls and mitigation options. *Nutr. Cycling Agroecosyst.* **1997**, *48*, (1), 139-153.



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